# Understanding Influence of Thermal History and Glass Chemistry on Kinetics of Phase Separation and Crystallization in Borosilicate Glass-Ceramic Waste Forms for Aqueous Reprocessed High Level Waste

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**Collaborators**:

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**ABSTRACT:**

An innovative glass-ceramic waste form is being developed by the US DOE to immobilize non-fissionable waste streams of alkali/alkaline-earths, lanthanides, and transition metals generated by the projected TRUEX$^{plus}$ process. Fundamental mechanistic understanding of thermodynamic phase transitions and crystallization mechanisms occurring during synthesis of this glass ceramic (as a function of thermal history and glass chemistry) is of paramount importance for international waste management programs in the US and UK, which seek to exploit glass-ceramic technology. The overarching goals of the project are to understand: (1) fundamental mechanisms of the *phase transitions* leading to the conversion of melt to multiphase glass ceramic as a function of composition and cooling rate; and (2) influence of *glass chemistry* on kinetics of phase separation and crystallization. These objectives will be realized through the performance of three inter-related tasks (i) Understanding kinetics of phase separation and crystallization in baseline glasses, (ii) Influence of glass chemistry on phase transitions and chemical durability, and (iii) *In-situ* high temperature studies. This project will provide much-needed fundamental science on the kinetics and mechanisms of liquid-liquid phase separation and crystallization of complex multi-phase glass ceramics for immobilization of mixed alkali/alkaline earth, lanthanide, and transition metals.

Major outcomes of this effort include: training of graduate students in glass ceramic engineering of nuclear waste forms; highly relevant publications in peer-reviewed journals; and formal technical exchange between US and UK researchers. Nuclear waste management is a long-term problem involving a worldwide technical community, and these projects solidify international cooperation towards joint technical goals. Consequently, this program is a joint collaborative enterprise between leading researchers from the US and UK who, collectively, bring mutually complementary and compatible skills, capabilities, and interests required to achieve a paradigm shift in technical understanding of crystallization in glass ceramics for radioactive waste immobilization. The team comprises researchers from two US universities (WSU and Rutgers), two UK universities (UoS and UoW), and a US national lab partner (PNNL). Prof. McCloy (WSU) has extensive experience with ceramic materials and spectroscopy, as well as managing complex technical projects. Prof. Goel (Rutgers) is an expert in thermal analysis of glasses and glass ceramics. UoS personnel have strong expertise in ceramic waste forms (Prof. Hyatt) and glasses of all types (Prof. Hand) as well as geochemical considerations of waste form durability (Dr. Corkhill) and multiscale modeling (Dr. Travis). Dr. Hanna (UoW) is a world expert on nuclear magnetic resonance, including difficult isotopes such as $^{43}$Ca, $^{95}$Mo, and $^{133}$Cs. The US portion of the research will be funded by the Department of Energy–Nuclear Energy NEUP program, and the UK portion will be funded by Engineering and Physical Sciences Research Council, Research Councils UK Energy Programme.